

Rayleigh Instability in Liquid Crystalline Jet

Leonid G. Fel and Yoram Zimmels

Department of Civil Engineering, Technion,
Haifa 32000, Israel

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Abstract

The capillary instability of liquid crystalline (LC) jets is considered in the framework of linear hydrodynamics of uniaxial nematic LC. The free boundary conditions of the problem are formulated in terms of mean surface curvature \mathcal{H} and Gaussian surface curvature \mathcal{G} . The static version of capillary instability is shown to depend on the elasticity modulus K , surface tension σ_0 , and radius r_0 of the LC jet, as expressed by the characteristic parameter $\varkappa = K/\sigma_0 r_0$. The problem of capillary instability in LC jets is solved exactly and a dispersion relation, which reflects the effect of elasticity, is derived. It is shown that increase of the elasticity modulus results in a decrease of both the cut off wavenumber k and the disturbance growth rate s . This implies enhanced stability of LC jets, compared to ordinary liquids. In the specific case, where the hydrodynamic and orientational LC modes can be decoupled, the dispersion equation is given in closed form.

Key words: Jet, Rayleigh Instability, Nematic Liquid Crystal

1 Introduction

The breakup of liquid jets, that are injected through a circular nozzle into stagnant fluids, has been the subject of widespread research over the years. Previous studies that followed the seminal works of Lord Rayleigh have established that the complex jet flow is influenced by a large number of parameters. These include nozzle internal flow effects, the jet velocity profile and the physical state of both liquid and gas. Notwithstanding the fact that the hydrodynamic equations are nonlinear, the linear stability theory can provide qualitative descriptions of breakup phenomena and predict the existence of different breakup regimes.

Rayleigh showed [1] by using a linear theory that the jet breakup is a consequence of hydrodynamic instability, or more exactly *capillary instability*. Neglecting the effect of the ambient fluid, the viscosity of the jet liquid, and gravity, he demonstrated that a cylindrical liquid jet is unstable with respect to disturbances characterized by wavelengths larger than the jet circumference. He also considered the case of a viscous jet in an inviscid gas and an inviscid gas jet in an inviscid liquid [2]. Weber [3] generalized Rayleigh's result for the case of a *Newtonian* viscous liquid and showed that the viscosity tends to reduce the breakage rate and increase the drop size. Chandrasekhchar [4] considered the effect of a uniform magnetic field on the capillary instability of a liquid jet. A mechanism of bending disturbances and of buckling, slowly moving, highly viscous jets, was presented by Taylor [5]. Further developments of the theory in Newtonian liquids was concerned with additional factors such as the dynamic action of the ambient gas (leading to atomization of the jet), the nonlinear interaction of growing modes that lead to satellite drop formation, and the spatial character of instability (see [6], [7]).

The capillary instability in jets, comprised of *non-Newtonian* suspensions and emulsions, presents a different category of cases which are governed by power-law (pseudoplastic and dilatant) liquids. The effective viscosity of the pseudoplastic liquid decreases with growth of strain rate, whereas in dilatant liquids, it increases [7]. The behaviour of capillary jets of dilute and concentrated polymer solutions suggest a strong influence of the macromolecular coils on their flow patterns [7]. Free jets of polymeric liquids, that exhibit oscillations, are reported in [8].

Recently the idea of Rayleigh instability was applied to tubular membranes in dilute ly-

otropic phases [9]. Their relaxation, following optical excitation, is characterized by a long time, and can be described by means of hydrodynamic approach [10]. Bending deformations of such membranes are governed by the Helfrich energy [11] which depends on the curvature of the tube. Thus, competition between the surface tension and curvature energy of the water immersed membrane renders the initial shape of the tube unstable. The hydrodynamic formalism used in [10] and the hydrodynamics of fluids with inner order such as liquid crystal (LC) [12] have similar features. In [10] the order parameter stands for a unit vector normal to the membrane surface. In contrast, the order parameter Q of a LC fluid, is defined throughout the space it occupies.

The continuum theory of LC phases has emerged as a rigorous part of condensed matter theory. The hydrodynamics of the LC phases was developed during the 70–80th and its predictions were successfully confirmed in many experimental observations. The combination of viscous and elastic properties is likely to produce new evolution patterns of hydrodynamic instabilities, in the context of Benard–Rayleigh, Marangoni and electrohydrodynamic effects [13], which cannot occur in ordinary liquids. However, its capillary instability, when in the form of a jet, was not considered as yet. In particular we refer to the *uniaxial nematic* phase.

The instability of a LC jet poses an additional challenge with respect to the effects listed above. This applies already within the framework of linear stability theory. The LC class of fluids seems to provide a good example of unique properties, as compared to polymer solutions. The elastic properties of a LC are expected to change the evolution patterns of jets which are made from them. In this work we derive a rigorous mathematical model of capillary instability for isothermal incompressible nematic LC jets. This model shows how the combined viscous and elastic properties of LC fluids determine the boundary conditions at the free surface, and the range where instability prevails.

2 Hydrodynamics of a liquid crystalline jet

In this Section, we formulate first the problem of capillary instability and then derive the basic equations which govern the linear hydrodynamics of a liquid crystalline jet. The flow of a nematic LC is described by a set of differential equations supplemented by boundary conditions on the LC free surface: continuity equation, Navier–Stokes equation of visco–elastic

LC, and Leslie–Ericksen equation of angular motion of the director $\mathbf{n}(\mathbf{r}, t)$.

The basic notations and linear hydrodynamic equations of uniaxial nematic liquid crystals follow the theory given in [12], [14], [15].

2.1 Basic notations and variables

The following basic variables describe the nematic LC medium: velocity $\mathbf{V}(\mathbf{r}, t)$, pressure $P(\mathbf{r}, t)$ and LC-director $\mathbf{n}(\mathbf{r}, t)$. The initial values of the functions will be denoted by "o", either as a subscript or superscript. The following notations, which are commonly accepted in the theory of LCs, are used henceforth:

1. The free energy density E_d of deformed non-chiral uniaxial nematic LC, given in quadratic approximation in terms of the derivatives $\partial\mathbf{n}/\partial x_j$ reads

$$2E_d = K_1 \operatorname{div}^2 \mathbf{n} + K_2 \langle \mathbf{n}, \operatorname{rot} \mathbf{n} \rangle^2 + K_3 [\mathbf{n} \times \operatorname{rot} \mathbf{n}]^2, \quad (1)$$

where $\langle \mathbf{a}, \mathbf{b} \rangle$ and $[\mathbf{a} \times \mathbf{b}]$ denote scalar and vector products of vectors, and $K_i \geq 0, i = 1, 2, 3$ are known as the Frank elasticity moduli. In the vicinity of a phase transition $K_i \propto Q^2$ [13] and in the isotropic phase they vanish.

2. The bulk molecular field \mathbf{F} and the Ericksen elastic stress tensor τ_{ki} , which set the equilibrium distribution of the \mathbf{n} -field in a LC, are determined by the following variational derivatives ¹

$$\mathbf{F} = \mathbf{M} - \mathbf{n} \langle \mathbf{n}, \mathbf{M} \rangle, \quad \text{or} \quad F_i = (\delta_{ij} - n_i n_j) M_j, \quad (2)$$

where

$$M_i = \frac{\partial}{\partial x_k} \frac{\partial E_d}{\partial (\partial_k n_i)} - \frac{\partial E_d}{\partial n_i}, \quad \tau_{ki} = \frac{\partial E_d}{\partial (\partial_k n_i)}, \quad \partial_k = \frac{\partial}{\partial x_k}, \quad (3)$$

i.e.

$$\begin{aligned} \mathbf{M} &= K_1 \operatorname{grad} \operatorname{div} \mathbf{n} - K_2 \{ \langle \mathbf{n}, \operatorname{rot} \mathbf{n} \rangle \operatorname{rot} \mathbf{n} + \operatorname{rot} (\langle \mathbf{n}, \operatorname{rot} \mathbf{n} \rangle \mathbf{n}) \} + \\ &\quad K_3 \{ \operatorname{rot} [\mathbf{n} \times [\mathbf{n} \times \operatorname{rot} \mathbf{n}]] + [[\mathbf{n} \times \operatorname{rot} \mathbf{n}] \times \operatorname{rot} \mathbf{n}] \}, \\ \tau_{ki} &= K_1 \delta_{ki} \operatorname{div} \mathbf{n} + K_2 \langle \mathbf{n}, \operatorname{rot} \mathbf{n} \rangle n_m \epsilon_{mki} + K_3 [[\mathbf{n} \times \operatorname{rot} \mathbf{n}] \times \mathbf{n}]_m \epsilon_{mki}, \end{aligned} \quad (4)$$

¹Here and throughout the paper, unless noted otherwise, we apply the summation rule over indices which are repeated in a tensor product, e.g. $a_{ij}b_{jk} = \sum_j a_{ij}b_{jk}$.

ϵ_{mki} is a completely antisymmetric unit tensor of the 3rd rank (Levi–Civita tensor).

3. If the deviations of the director $\mathbf{n} = \mathbf{n}^0 + \mathbf{n}^1$ from its initial orientation \mathbf{n}^0 are small, then

$$n_x^0 = n_y^0 = 0, \quad n_z^0 = 1, \quad 1 \gg n_x^1, n_y^1 \gg n_z^1 \sim (n_x^1)^2, (n_y^1)^2, \quad (5)$$

and simple algebra yields the following linear approximation

$$M_x = \hat{K}n_x^1 + (K_1 - K_2) \frac{\partial^2 n_y^1}{\partial x \partial y}, \quad M_y = \hat{K}n_y^1 + (K_1 - K_2) \frac{\partial^2 n_x^1}{\partial x \partial y}, \quad M_z = (K_1 - K_3) \frac{\partial}{\partial z} \text{div } \mathbf{n}^1, \quad (6)$$

where $\hat{K} = K_1 \frac{\partial^2}{\partial x^2} + K_2 \frac{\partial^2}{\partial y^2} + K_3 \frac{\partial^2}{\partial z^2}$, and by virtue of (2), $F_x = M_x$, $F_y = M_y$, $F_z = 0$. If further simplification through single elastic approximation $K_1 = K_2 = K_3 = K$ is applied, then

$$F_x = K \Delta_3 n_x^1, \quad F_y = K \Delta_3 n_y^1, \quad F_z = 0, \quad \Delta_3 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}, \quad (7)$$

where Δ_3 is the three-dimensional Laplacian. Similar considerations regarding the Ericksen stress tensor τ_{ki} give

$$\begin{aligned} \tau_{xx} = \tau_{yy} = \tau_{zz} &= K_1 \text{div } \mathbf{n}^1, \quad \tau_{xy} = -\tau_{yx} = K_2 \left(\frac{\partial n_y^1}{\partial x} - \frac{\partial n_x^1}{\partial y} \right), \\ \tau_{yz} = -\tau_{zy} &= K_3 \left(\frac{\partial n_z^1}{\partial y} - \frac{\partial n_y^1}{\partial z} \right), \quad \tau_{zx} = -\tau_{xz} = K_3 \left(\frac{\partial n_x^1}{\partial z} - \frac{\partial n_z^1}{\partial x} \right), \end{aligned} \quad (8)$$

The stresses given by (8) do not contribute to the non-dissipative stress tensor T_{ik}^r used in the linear hydrodynamics of LCs (see (9) below).

4. The reactive (non-dissipative) T_{ik}^r and dissipative T_{ik}^d stress tensors are defined as follows

$$T_{ik}^r = -P \delta_{ik} - \tau_{kj} \frac{\partial n_j}{\partial x_i} - \frac{\lambda}{2} (n_i F_k + n_k F_i) + \frac{1}{2} (n_i F_k - n_k F_i), \quad (9)$$

$$\begin{aligned} T_{ik}^d &= 2\eta_1 \Upsilon_{ik} + (\eta_2 - \eta_1) \delta_{ik} \text{div } \mathbf{V} + (\eta_1 - \eta_2 + \eta_4) (\delta_{ik} n_j \Upsilon_{jm} n_m + n_i n_k \text{div } \mathbf{V}) + \\ &(\eta_3 - 2\eta_1) (n_i \Upsilon_{kj} n_j + n_k \Upsilon_{ij} n_j) + (\eta_1 + \eta_2 + \eta_5 - 2\eta_3 - 2\eta_4) n_i n_k n_j n_m \Upsilon_{jm}, \end{aligned} \quad (10)$$

where the antisymmetric Ω_{ik} (*vorticity*) and symmetric Υ_{ik} parts of the derivative $\partial_k V_i$ read

$$\Omega_{ik} = \frac{1}{2} \left(\frac{\partial V_k}{\partial x_i} - \frac{\partial V_i}{\partial x_k} \right), \quad \Upsilon_{ik} = \frac{1}{2} \left(\frac{\partial V_k}{\partial x_i} + \frac{\partial V_i}{\partial x_k} \right), \quad (11)$$

Five independent viscous moduli η_j , kinetic coefficient λ , and rotational viscosity γ_1 , determine the dissipative stress tensor T_{ik}^d , the 4th-rank viscosity tensor η_{ikjm} , and the dissipative

function D in the absence of heat fluxes

$$\begin{aligned}
D &= \eta_{ikjm} \Upsilon_{ik} \Upsilon_{jm} + \frac{1}{\gamma_1} \mathbf{F}^2, \quad T_{ik}^d = \eta_{ikjm} \Upsilon_{jm}, \\
\eta_{ikjm} &= \eta_1 (\xi_{ij} \xi_{km} + \xi_{kj} \xi_{im}) + (\eta_2 - \eta_1) \xi_{ik} \xi_{jm} + \frac{\eta_3}{2} (n_i n_j \xi_{km} + n_k n_j \xi_{im} + n_i n_m \xi_{kj} + \\
&\quad + n_k n_m \xi_{ij}) + \eta_4 (n_i n_k \xi_{jm} + n_j n_m \xi_{ik}) + \eta_5 n_i n_k n_j n_m, \quad \xi_{ik} = \delta_{ik} - n_i n_k.
\end{aligned} \tag{12}$$

The tensor η_{ikjm} consists of five independent uniaxial invariants [12] and is highly symmetrical $\eta_{ikjm} = \eta_{kimj} = \eta_{jmik}$. The requirement that D be positive translates into,

$$\eta_1 \geq 0, \quad \eta_2 \geq 0, \quad \eta_3 \geq 0, \quad \eta_5 \geq 0, \quad \eta_2 \eta_5 \geq \eta_4^2, \quad \gamma_1 \geq 0. \tag{13}$$

The parameter λ is close to +1 or -1 for rod-like or disk-like molecules, respectively. If the liquid is visco-isotropic, then $\lambda = 0$.

5. The hydrodynamic reactive (non-dissipative) \mathbf{m}^r and dissipative \mathbf{m}^d fields are defined as follow

$$m_i^r = -\langle \mathbf{V}, \nabla_3 \rangle n_i + n_k \Omega_{ki} + \lambda \xi_{ij} \Upsilon_{jk} n_k, \quad \mathbf{m}^d = \frac{1}{\gamma_1} \mathbf{F}, \tag{14}$$

where ∇_3 is the three-dimensional gradient operator, $(\nabla_3)^2 = \Delta_3$.

6. The surface tension σ of nematic LC is given by [16],

$$\sigma = \sigma_0 + \sigma_1 \langle \mathbf{n}, \mathbf{e} \rangle^2, \tag{15}$$

where σ_0 and σ_1 are isotropic and anisotropic surface tension moduli respectively, and \mathbf{e} is a unit normal vector to the LC surface.

7. In the case of an incompressible LC ($\eta_1^{\text{in}} = \eta_2^{\text{in}}, \eta_4^{\text{in}} = 0$) the tensors T_{ik}^d and η_{ikjm} take the following form,

$$\begin{aligned}
T_{ik}^{\text{din}} &= 2\eta_1^{\text{in}} \Upsilon_{ik} + (\eta_3^{\text{in}} - 2\eta_1^{\text{in}}) (n_i \Upsilon_{kj} n_j + n_k \Upsilon_{ij} n_j) + (2\eta_1^{\text{in}} + \eta_5^{\text{in}} - 2\eta_3^{\text{in}}) n_i n_k n_j n_m \Upsilon_{jm} \\
\eta_{ikjm}^{\text{in}} &= \eta_1^{\text{in}} (\xi_{ij} \xi_{km} + \xi_{kj} \xi_{im}) + \frac{\eta_3^{\text{in}}}{2} (n_i n_j \xi_{km} + n_k n_j \xi_{im} + n_i n_m \xi_{kj} + n_k n_m \xi_{ij}) + \eta_5^{\text{in}} n_i n_k n_j n_m,
\end{aligned} \tag{16}$$

where "in" denotes incompressibility condition.

8. The first two terms in (10) and the second equation of (12) correspond to ordinary compressible liquids with isotropic invariance. The simplification of (10) results from

$$\eta_3^{\text{L}} = 2\eta_1^{\text{L}}, \quad \eta_4^{\text{L}} = \eta_2^{\text{L}} - \eta_1^{\text{L}}, \quad \eta_5^{\text{L}} = \eta_2^{\text{L}} + \eta_1^{\text{L}},$$

so that

$$T_{ik}^{\text{dl}} = 2\eta_1^{\text{L}}\Upsilon_{ik} + (\eta_2^{\text{L}} - \eta_1^{\text{L}}) \delta_{ik} \text{div} \mathbf{V} , \quad \eta_{ikjm}^{\text{L}} = \eta_1^{\text{L}}(\delta_{ij}\delta_{km} + \delta_{im}\delta_{kj}) + (\eta_2^{\text{L}} - \eta_1^{\text{L}})\delta_{ik}\delta_{jm} .$$

The coefficients η_1^{L} and $\eta_2^{\text{L}} - \eta_1^{\text{L}}$ are known as *the first* and *second* isotropic viscosities.

9. Another system of viscous moduli α_i (called *Lesli viscosities*) relate dissipative and kinetic moduli in the following way ²

$$\begin{aligned} \eta_1 &= \frac{\alpha_4}{2} , \quad \lambda = -\frac{\gamma_2}{\gamma_1} , \quad \eta_5 = \alpha_1 + \alpha_4 + \alpha_5 + \alpha_6 , \quad \gamma_1 = \alpha_3 - \alpha_2 , \quad \gamma_2 = \alpha_3 + \alpha_2 , \\ \eta_3 - 2\eta_1 &= \alpha_5 + \alpha_2\lambda , \quad 2\eta_1 + \eta_5 - 2\eta_3 = \alpha_1 + \frac{\gamma_2^2}{\gamma_1} , \end{aligned} \quad (17)$$

with the support of Onzager–Parodi relation [17] $\alpha_3 + \alpha_2 = \alpha_6 - \alpha_5$. In the vicinity of phase transition, the viscous moduli α_i have different dependences upon the order parameter Q : $\alpha_1 \propto Q^2, \alpha_2, \alpha_3, \alpha_5, \alpha_6 \propto Q, \alpha_4 \propto Q^0$ [13].

Tables 1 and 2 (see Appendix) summarize viscosities and other physical parameters that characterize the most frequently used and well studied nematic LC, also known as *MBBA* and *PAA*.

2.2 Basic equations

The complete system of hydrodynamic equations for nematic LC reflect the conservation laws of mass, and of linear and angular momenta.

1. Continuity equation

$$\frac{\partial \rho}{\partial t} + \text{div}(\rho \mathbf{V}) = 0 . \quad (18)$$

2. Navier–Stokes equation for visco–elastic LC

$$\rho \frac{\partial V_i}{\partial t} + \rho \langle \mathbf{V}, \nabla_3 \rangle V_i = \frac{\partial}{\partial x_k} (T_{ik}^{\text{r}} + T_{ik}^{\text{d}}) . \quad (19)$$

3. Lesli–Ericksen equation of angular motion of the director $\mathbf{n}(\mathbf{r}, t)$

$$\frac{\partial \mathbf{n}}{\partial t} = \mathbf{m}^{\text{r}} + \mathbf{m}^{\text{d}} . \quad (20)$$

²The correct expression for η_5 is given in [14].

The last equation is written for a negligible specific angular moment of inertia \mathcal{J}_{LC} of the LC, namely, $\mathcal{J}_{LC} \ll \rho r_0^2$, where r_0 is a characteristic size of the system. This is true in our case, where r_0 denotes radius of the jet.

Consider an isothermal incompressible jet, flowing along the z axis, out of a nozzle at a velocity \mathbf{V} . The initial orientation of director \mathbf{n}^0 is assumed collinear with \mathbf{V} . The deviation from initial values of the director and pressure are defined as $\mathbf{n}^1 = \mathbf{n} - \mathbf{n}^0$, and $P_1 = P - P_0$ respectively, where $P_0 = \sigma/r_0$ is the unperturbed pressure within the cylindrical jet. Applying the linear approximation $|\mathbf{n}^1| \ll 1$, equations (18)–(20) are simplified as follows

$$\begin{aligned} \operatorname{div} \mathbf{V} &= 0, \quad \rho \frac{\partial V_i}{\partial t} = -\frac{\partial P_1}{\partial x_i} + \frac{\partial T_{ik}^{\text{din}}}{\partial x_k} + \frac{1-\lambda}{2} n_i^0 \operatorname{div} \mathbf{F} - \frac{1+\lambda}{2} \langle \mathbf{n}^0, \nabla_3 \rangle F_i, \\ \frac{\partial n_i^1}{\partial t} &= n_k^0 \Omega_{ki} + \lambda \xi_{ij}^0 \Upsilon_{jk} n_k^0 + \frac{1}{\gamma_1} F_i, \quad \xi_{ij}^0 = \delta_{ij} - n_i^0 n_j^0, \quad i, j, k = x, y, z. \end{aligned} \quad (21)$$

Choosing $n_z^0 = 1$, gives $F_z = 0$ and hence

$$0 = \frac{\partial V_x}{\partial x} + \frac{\partial V_y}{\partial y} + \frac{\partial V_z}{\partial z}, \quad (22)$$

$$\rho \frac{\partial V_x}{\partial t} = -\frac{\partial P_1}{\partial x} + \left[\beta_1 \Delta_2 + \beta_2 \frac{\partial^2}{\partial z^2} \right] V_x + (\beta_2 - \beta_1) \frac{\partial^2 V_z}{\partial x \partial z} - \frac{\lambda+1}{2} \frac{\partial F_x}{\partial z}, \quad (23)$$

$$\rho \frac{\partial V_y}{\partial t} = -\frac{\partial P_1}{\partial y} + \left[\beta_1 \Delta_2 + \beta_2 \frac{\partial^2}{\partial z^2} \right] V_y + (\beta_2 - \beta_1) \frac{\partial^2 V_z}{\partial y \partial z} - \frac{\lambda+1}{2} \frac{\partial F_y}{\partial z},$$

$$\rho \frac{\partial V_z}{\partial t} = -\frac{\partial P_1}{\partial z} + \left[\beta_2 \Delta_2 + \beta_3 \frac{\partial^2}{\partial z^2} \right] V_z - \frac{\lambda-1}{2} \operatorname{div} \mathbf{F},$$

$$\frac{\partial n_x^1}{\partial t} = \frac{\lambda+1}{2} \frac{\partial V_x}{\partial z} + \frac{\lambda-1}{2} \frac{\partial V_z}{\partial x} + \frac{F_x}{\gamma_1}, \quad (24)$$

$$\frac{\partial n_y^1}{\partial t} = \frac{\lambda+1}{2} \frac{\partial V_y}{\partial z} + \frac{\lambda-1}{2} \frac{\partial V_z}{\partial y} + \frac{F_y}{\gamma_1}, \quad \frac{\partial n_z^1}{\partial t} = 0,$$

where $\Delta_2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$ is the two-dimensional Laplacian, $\beta_1 = \eta_1^{\text{in}}$, $\beta_2 = \eta_3^{\text{in}}/2$, $\beta_3 = \eta_5^{\text{in}} - \eta_3^{\text{in}}/2$ and F_x, F_y are given in (7). As isotropic viscosity means $\beta_i = \beta$, the above mentioned liquid crystals, *MBBA* and *PAA*, are clearly far from being isotropic (see Tables 1, 2 in Appendix).

In order to make the problem more specific and easier to solve, we consider *axisymmetrical* disturbances in a system of cylindrical LC jet, with radius r_0 and subject to the single elastic approximation ($K_i = K$). This provides the simplest approximation which still preserves the influence of elastic forces, on the hydrodynamics of an incompressible and elastic LC. In this

case

$$0 = \frac{\partial V_z}{\partial z} + \frac{\partial V_r}{\partial r} + \frac{V_r}{r}, \quad (25)$$

$$\rho \frac{\partial V_r}{\partial t} = -\frac{\partial P_1}{\partial r} + \left[\beta_1 \left(\Delta_{2c} - \frac{1}{r^2} \right) + \beta_2 \frac{\partial^2}{\partial z^2} \right] V_r + (\beta_2 - \beta_1) \frac{\partial^2 V_z}{\partial r \partial z} - \mu_1 \frac{\partial F_r}{\partial z}, \quad (26)$$

$$\rho \frac{\partial V_z}{\partial t} = -\frac{\partial P_1}{\partial z} + \left[\beta_2 \Delta_{2c} + \beta_3 \frac{\partial^2}{\partial z^2} \right] V_z - \mu_2 \left(\frac{\partial F_r}{\partial r} + \frac{F_r}{r} \right), \quad (27)$$

$$\gamma_1 \frac{\partial n_r^1}{\partial t} = \gamma_1 \mu_1 \frac{\partial V_r}{\partial z} + \gamma_1 \mu_2 \frac{\partial V_z}{\partial r} + F_r, \quad n_z^1 = 0, \quad (28)$$

where

$$\Delta_{2c} = \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r}, \quad F_r = K \left(\Delta_{2c} - \frac{1}{r^2} + \frac{\partial^2}{\partial z^2} \right) n_r^1, \quad \mu_1 = \frac{\lambda + 1}{2}, \quad \mu_2 = \frac{\lambda - 1}{2}. \quad (29)$$

Equations (25)–(28) describe ordinary linear hydrodynamic behaviour of isotropic incompressible liquids provided that the LC properties vanish: $K, \gamma_1 \rightarrow 0$ and $\beta_i = \beta$. The result is the well known continuity and linearized Navier–Stokes equations.

$$\text{div} \mathbf{V} = 0, \quad \rho \frac{\partial \mathbf{V}}{\partial t} = -\nabla P_1 + \beta \Delta_3 \mathbf{V}. \quad (30)$$

2.3 Boundary conditions at free surface

Boundary conditions at the free surface of a liquid crystal state that the jump in normal stress consists of two parts: one depends on the surface tension σ , and the other on the elastic disturbance W_{elast} of the uniform director field $\mathbf{n}_0(\mathbf{r})$. Assuming that no tangential stresses exist at the free surface, the boundary conditions can be expressed as,

$$(T_{ik}^r + T_{ik}^{\text{din}}) e_k + (2\sigma \mathcal{H} + W_{\text{elast}}) e_i + \frac{\partial \sigma}{\partial x_i} = 0 \quad \text{at} \quad r = r_0, \quad (31)$$

where e_i are the components of the normal unit vector \mathbf{e} in the reference frame of the LC–cylinder, and $\mathcal{H} = 1/2 (1/R_1 + 1/R_2)$ denotes mean surface curvature with principal radii R_1 and R_2 .

The non–hydrodynamic part of boundary conditions at the free surface holds, provided that the scale of deformation of the initial surface is considerably larger compared to the molecular length of LCs³. This dictates a tangential behaviour of a smoothly disturbed

³Strictly speaking, this assumption is correct when an equilibrium distribution of director field $\mathbf{n}(\mathbf{r})$ is free of singularities. The problem of *minimal surface of LC drop* presents another situation wherein an essential rearrangement of the field $\mathbf{n}(\mathbf{r})$, at the surface, can diminish the total energy by destroying the disclination core within the drop.

director \mathbf{n} at the free surface, $e_z \ll e_r \sim 1$:

$$\langle \mathbf{e}, \mathbf{n} \rangle = 0 \quad \rightarrow \quad e_z + n_r^1 = 0 \quad \text{at} \quad r = r_0 . \quad (32)$$

The last constraint cancels the gradient term in (31). Finally we come to the boundary conditions in the linear approximation of the variables n_r^1, V_r, V_z, P_1

$$T_{rr}^r + T_{rr}^{\text{din}} + 2\sigma\mathcal{H} + W_{\text{elast}} = 0, \quad T_{zr}^r + T_{zr}^{\text{d}} = 0 . \quad (33)$$

Substitution of the expressions for the reactive and dissipative stress tensors gives

$$2\beta_1 \Upsilon_{rr} - P_1 = 2\sigma_0 (\mathcal{H}_0 - \mathcal{H}) - W_{\text{elast}}, \quad 2\beta_2 \Upsilon_{zr} = \mu_2 F_r \quad \text{at} \quad r = r_0 . \quad (34)$$

where $\mathcal{H}_0 = 1/2r_0$ is the initial mean curvature of the LC-cylinder. The equations for a jet surface, disturbed by a wave $\zeta(z, t)$, and its radial velocity $\partial\zeta/\partial t$, are given by

$$r(z, t) = r_0 + \zeta(z, t), \quad V_r = \frac{\partial\zeta}{\partial t} \quad \text{at} \quad r = r_0 , \quad (35)$$

where $\zeta \ll r_0$ is the radial displacement of a surface point. The principal radii of the surface curvature, in the context of linear approximation with respect to ζ , and its derivatives can be expressed as,

$$\frac{1}{R_1} = \frac{1}{r_0 + \zeta} \cong \frac{1}{r_0} - \frac{\zeta}{r_0^2}, \quad \frac{1}{R_2} \cong -\frac{\partial^2 \zeta}{\partial z^2} . \quad (36)$$

This transforms the boundary conditions (32), (34) into

$$n_r^1 = \frac{\partial\zeta}{\partial z}, \quad V_r = \frac{\partial\zeta}{\partial t}, \quad (37)$$

$$2\beta_2 \Upsilon_{zr} = \mu_2 F_r, \quad (38)$$

$$P_1 - 2\beta_1 \Upsilon_{rr} = -\sigma_0 \left(\frac{\zeta}{r_0^2} + \frac{\partial^2 \zeta}{\partial z^2} \right) + W_{\text{elast}} . \quad (39)$$

The term W_{elast} deserves further discussion. It reflects the existence of normal stresses, at the surface, which arise due to the resistance of the uniformly orientated continuous LC media to a surface disturbance. W_{elast} vanishes in undisturbed LC jets and it depends linearly on the elastic modulus K , radius r_0 and derivatives of ζ . Moreover, an invariance of the problem with respect to inversion of the z -axis requires sole dependence on derivatives of even orders. An explicit expression for W_{elast} is derived in Section 3.1.

3 Plateau instability in a LC cylinder

Before proceeding to tackle the sophisticated mathematics of equations (25)–(28), as supplemented by boundary conditions (37)–(39), capillary instability of the LC cylinder is discussed. This is done by applying the Plateau considerations [18] on the figures of a liquid mass withdrawn from the action of gravity.

Consider a LC cylinder with a surface disturbed as specified by (35), where $\zeta = \zeta_0 \cos kz$, ζ_0 is small compared to r_0 , and $k = 2\pi/\Lambda$, Λ being the disturbance *wavelength*. The idea of Plateau, applied here, is to find such *cut-off wavelength* Λ_s of the disturbance, that defines breakage of the cylinder into droplets with due decrease of the total energy.

The volume v enclosed within one wave length is given by

$$v = \int_v dv = \pi \left(r_0^2 + \frac{1}{2} \zeta_0^2 \right) \rightarrow r_0 = \sqrt{\frac{v}{\pi}} \left(1 - \frac{1}{4} \frac{\pi \zeta_0^2}{v} \right), \quad (40)$$

where r_0 in the right h.s. of (40) is given as a second order expansion ζ_0 . The total energy \mathcal{E} of the LC cylinder with a disturbed director field $\mathbf{n}(\mathbf{r})$ is given by

$$\mathcal{E} = \sigma_0 \int_s ds + \frac{K}{2} \int_v (\text{div}^2 \mathbf{n} + \text{rot}^2 \mathbf{n}) dv. \quad (41)$$

The static director field $\mathbf{n}(\mathbf{r})$ can be found from equation (29) and the attendant boundary condition (37)

$$n_z^0 = 1, \quad F_r = 0 \rightarrow \left(\Delta_{2c} - \frac{1}{r^2} + \frac{\partial^2}{\partial z^2} \right) n_r^1 = 0, \quad n_r^1 = \frac{\partial \zeta}{\partial z} \quad \text{at} \quad r = r_0. \quad (42)$$

Equation (42) is satisfied by the following solution, which is finite at $r = 0$

$$n_r^1(r, z) = -\frac{k\zeta_0}{I_1(kr_0)} I_1(kr) \sin kz, \quad (43)$$

where $I_m(x)$ is a modified Bessel function of order m . The contribution of elastic forces is determined by

$$\text{div}^2 \mathbf{n} + \text{rot}^2 \mathbf{n} = k^2 \left[\frac{k\zeta_0}{I_1(kr_0)} \right]^2 [A_1^2(kr) \sin^2 kz + A_2^2(kr) \cos^2 kz] \quad (44)$$

where

$$A_1(y) = \frac{dI_1(y)}{dy} + \frac{1}{y} I_1(y), \quad A_2(y) = I_1(y).$$

A simple integration of (41) gives

$$\mathcal{E} = 2\pi\sigma_0 r_0 \left(1 + \frac{1}{4} k^2 \zeta_0^2 \right) + \frac{\pi}{2} K \left[\frac{k\zeta_0}{I_1(kr_0)} \right]^2 \int_0^{kr_0} [A_1^2(y) + A_2^2(y)] y dy. \quad (45)$$

Inserting r_0 from (40) into the first term above, we obtain

$$\mathcal{E} - 2\sigma_0\sqrt{\pi v} = \sigma_0 \frac{\pi\zeta_0^2}{2r_0} (\varpi^2 - 1) + \frac{\pi K}{2} \left[\frac{\zeta_0\varpi}{r_0 I_1(\varpi)} \right]^2 \int_0^\varpi [A_1^2(y) + A_2^2(y)] y dy, \quad \varpi = kr_0. \quad (46)$$

The positive root $\varpi_s = k_s r_0$, given in the right h.s. of (46), determines the cut-off wavelength Λ_s of capillary disturbances, which renders the LC cylinder unstable. Subsequent disintegration into detached masses is favored by the decrease in \mathcal{E}

$$(\varpi_s^2 - 1) + \varkappa \frac{\varpi_s^2}{I_1^2(\varpi_s)} \int_0^{\varpi_s} [A_1^2(y) + A_2^2(y)] y dy = 0, \quad \varkappa = \frac{K}{\sigma_0 r_0}, \quad (47)$$

where the subscript "s" denotes the static nature of Plateau instability.

The quadratic approximation (1) with respect to the derivatives $\partial \mathbf{n} / \partial x_j$, which provides the basis for the Frank theory, makes the expression (47) correct only in terms of the ϖ_s^2 approximation. Indeed, the power of ϖ_s in (47) should not exceed 2, otherwise the calculation becomes inconsistent. Thus, we get

$$\mathcal{E} - 2\sigma_0\sqrt{\pi v} = \sigma_0 \frac{\pi\zeta_0^2}{2r_0} (\varpi^2 - 1) + \pi K k^2 \zeta_0^2 \quad \text{and} \quad \varpi_s = \frac{1}{\sqrt{1 + 2\varkappa}}. \quad (48)$$

The asymptotic behaviour of $\varpi_s(\varkappa)$ shows two important limits:

$$\varpi_s = 1 - \varkappa \quad \text{if} \quad \varkappa \ll 1; \quad \varpi_s = \frac{1}{\sqrt{2\varkappa}} \left(1 - \frac{1}{4\varkappa} \right) \quad \text{if} \quad \varkappa \gg 1. \quad (49)$$

Figure 1 shows a plot of $k_s r_0$ vs. \varkappa for Plateau instabilities in LC and in ordinary liquid.

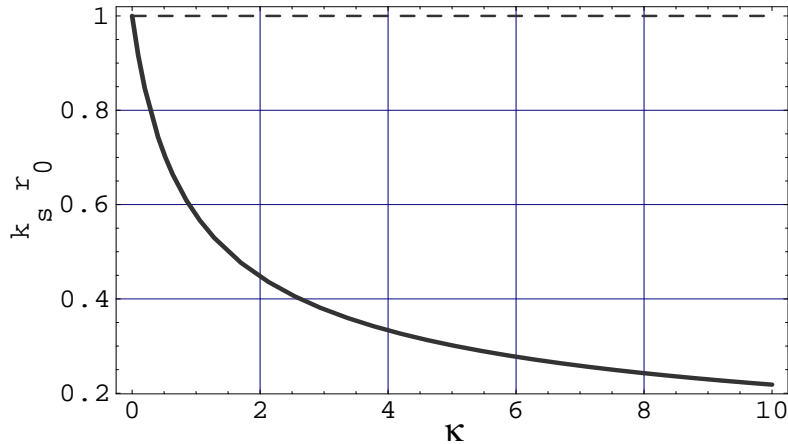


Figure 1: Universal plots of $k_s r_0$ vs. \varkappa for Plateau instabilities in LC cylinder (*plain line*), and in ordinary liquid $k_s r_0 = 1$ (*dashed line*).

The corresponding asymptotic cut-off wavelength Λ_s are obtained as

$$\Lambda_s = 2\pi r_0 (1 + \varkappa) \quad \text{if} \quad \varkappa \ll 1; \quad \Lambda_s = 2\pi \sqrt{\frac{2K}{\sigma_0}} \sqrt{r_0} \left(1 + \frac{1}{4\varkappa} \right) \quad \text{if} \quad \varkappa \gg 1. \quad (50)$$

This result shows that $k \geq k_s$ increases the total energy \mathcal{E} of the disturbed system, whereas $k \leq k_s$ decreases it. According to (49), there are two marginal regimes of instability:

- *Capillary regime* $r_0 \gg K/\sigma_0$. Here Λ_s is close in value to the circumference of the cylinder and the elastic deformation contribution $\int E_d dv$, to the total energy \mathcal{E} , is negligible. This regime must apply to a wide range of nematic LC, since the common values of $K \simeq 10^{-11} J/m$ [13] and $\sigma_0 \simeq 10^{-2} J/m^2$ [20] lead to $K/\sigma_0 \simeq 10^{-9} m$. This value is evidently smaller than the presently attainable radii of the jet.

- *Elastic regime* $r_0 \ll K/\sigma_0$. This case reflects the dominance of elastic deformation and predicts an unusual behaviour for $\Lambda_s \sim \sqrt{r_0}$.

This regime cannot be reached by simple increase of the elastic moduli since their magnitude is determined by $K \sim \kappa T/a$, where $\kappa T \sim 10^{-20} J$ is the Boltzmann thermal energy at room temperature, and $a \sim 10^{-9} m$ denotes molecular length of LC. In contrast, the effect of surface tension can be diminished by surfactants or by charging the surface of the liquid. In the latter case the charge can virtually eliminate the effect of surface tension and provide the conditions where the elastic forces predominate.

3.1 W_{elast} and Gaussian surface curvature

The straightforward way to derive an expression for W_{elast} is to solve the elastic problem for the stresses existing on a deformed axisymmetric surface of a LC cylinder. This relates to the Plateau instability, which obviates the need to repeat the entire procedure.

When we turn from Plateau considerations on the *static* instability of LC cylinders to the capillary instability of LC jets, the question is whether the cut-off wavelengths of both the static Λ_s and hydrodynamic Λ_d problems coincide. This question was skipped by Rayleigh in his studies of isotropic viscous liquids, since for ordinary liquids both cut-off wavelengths always coincide $\Lambda_s \equiv \Lambda_d$. This identity reflects a deep equivalence principle of the bifurcation point for non-trivial steady state of dynamic system, and the threshold of static instability concerned with a minimum of its free energy \mathcal{E} [4].

Making use of $\Lambda_s \equiv \Lambda_d$ we construct the term W_{elast} which enters the boundary condition (39). To this end, we examine and represent the total energy (48) as follows

$$\mathcal{E} - 2\sigma_0\sqrt{\pi v} = \frac{\pi\zeta_0 r_0}{2} \left[-\sigma_0 \left(\frac{\zeta_0}{r_0^2} - \zeta_0 k^2 \right) + 2K \frac{\zeta_0}{r_0} k^2 \right]. \quad (51)$$

Next, we compare the expression within the brackets with the right h.s. of (39). This gives W_{elast} , which generates the elastic contribution in (51)

$$W_{elast} = 2K\mathcal{G}, \quad \mathcal{G} = \frac{1}{R_1 R_2} = -\frac{1}{r_0} \frac{\partial^2 \zeta}{\partial z^2}, \quad (52)$$

where \mathcal{G} is the Gaussian surface curvature in accordance with (36). Thus the final expression for boundary conditions (31) is based on two fundamental invariants of the surface curvature, i.e. mean surface curvature \mathcal{H} , and Gaussian surface curvature \mathcal{G} .

4 Dispersion relation

Rayleigh was the first to observe [1] that contrary to Plateau, the instability problem is not so definite. The mode whereby a system deviates from unstable equilibrium must depend on the nature and characteristics of the small displacements to which this system is subjected. In the absence of such displacement, any system, however unstable, cannot depart from equilibrium. These characteristics, being hydrodynamic, reflect the effect of viscosity, which predominates over that of inertia. In the case of ordinary liquids, the mode of *maximum* instability which corresponds to the wavelength $\Lambda_R = 4.508 \times 2r_0$ exceeds the circumference of the liquid cylinder. We anticipate that the instability of LC jets possesses similar features.

The fact that a velocity potential does not exist in an anisotropic visco-elastic liquid, dictates a standard approach to this problem which was elaborated first by Rayleigh [2]. Let us define the *Stokes stream function* $\Psi(\mathbf{r}, t)$ and a *director potential* $\Theta(\mathbf{r}, t)$ as,

$$V_r = -\frac{1}{r} \frac{\partial \Psi}{\partial z}, \quad V_z = \frac{1}{r} \frac{\partial \Psi}{\partial r} \quad \text{and} \quad n_r^1 = \frac{\partial \Theta}{\partial r}, \quad (53)$$

so that the continuity equation (25) holds. From the other three equations (26)–(28) we have

$$\frac{\partial P_1}{\partial r} = (\beta_2 - \beta_1) \frac{\partial^2}{\partial r \partial z} \left(\frac{1}{r} \frac{\partial \Psi}{\partial r} \right) - \frac{1}{r} \frac{\partial}{\partial z} \left[\beta_1 r \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \Psi}{\partial r} \right) + \beta_2 \frac{\partial^2 \Psi}{\partial z^2} - \rho \frac{\partial \Psi}{\partial t} + \mu_1 r F_r \right] \quad (54)$$

$$\frac{\partial P_1}{\partial z} = \frac{1}{r} \frac{\partial}{\partial r} \left[\beta_2 r \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \Psi}{\partial r} \right) + \beta_3 \frac{\partial^2 \Psi}{\partial z^2} - \rho \frac{\partial \Psi}{\partial t} - \mu_2 r F_r \right], \quad (55)$$

$$\frac{\partial^2 \Theta}{\partial r \partial t} = \frac{1}{r} \left[\mu_2 r \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \Psi}{\partial r} \right) - \mu_1 \frac{\partial^2 \Psi}{\partial z^2} \right] + \frac{1}{\gamma_1} F_r, \quad F_r = K \left(\Delta_{2c} + \frac{\partial^2}{\partial z^2} - \frac{1}{r^2} \right) \frac{\partial \Theta}{\partial r}. \quad (56)$$

Applying the commutation rules give,

$$\left(\Delta_{2c} - \frac{1}{r^2} \right) \frac{\partial \Theta}{\partial r} = \frac{\partial}{\partial r} \Delta_{2c} \Theta \quad \rightarrow \quad F_r = K \frac{\partial}{\partial r} \left(\Delta_{2c} + \frac{\partial^2}{\partial z^2} \right) \Theta,$$

which facilitates simplification of the above equations. Assuming that an axisymmetrical disturbance, characterized by a wavelength $2\pi/k$, increases exponentially in time with the growth rate s , gives,

$$\{\Psi, \Theta, \zeta, P_1, F_r\} = \{i\psi(r), i\theta(r), \varsigma(r), p(r), if(r)\} \times e^{st+ikz}, \quad (57)$$

Inserting (57) into (54)–(56) gives rise to the following amplitude equations

$$\frac{1}{k} \frac{\partial p}{\partial r} = \beta_4 \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \psi}{\partial r} \right) - (\beta_2 k^2 + s\rho) \frac{\psi}{r} + \mu_1 f, \quad \beta_4 = 2\beta_1 - \beta_2, \quad (58)$$

$$kp = \frac{1}{r} \frac{\partial}{\partial r} \left\{ r \left[\beta_2 \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \psi}{\partial r} \right) - (\beta_3 k^2 + s\rho) \frac{\psi}{r} - \mu_2 f \right] \right\}, \quad (59)$$

$$s \frac{\partial \theta}{\partial r} = \mu_2 \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \psi}{\partial r} \right) + \mu_1 k^2 \frac{\psi}{r} + \frac{1}{\gamma_1} f, \quad f = K \frac{\partial}{\partial r} (\Delta_{2c} - k^2) \theta, \quad (60)$$

The new variables in (57) require reformulation of the boundary conditions (37)–(39) as follows,

$$k\varsigma = \frac{\partial \theta}{\partial r}, \quad s\varsigma = k \frac{\psi}{r}, \quad \frac{\mu_2}{\beta_2} f = \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \psi}{\partial r} \right) + k^2 \frac{\psi}{r}, \quad p = 2\beta_1 k \frac{\partial}{\partial r} \left(\frac{\psi}{r} \right) + \varsigma \Gamma \quad (61)$$

where

$$\Gamma = \sigma_0 \left(k^2 - \frac{1}{r_0^2} \right) + 2K \frac{1}{r_0} k^2.$$

The real form of the amplitude equations (58)–(60) and boundary conditions (61) imply that (57) divides the five variables into two groups: P_1, ζ and Ψ, Θ, F_r . These groups are shifted with respect to each other by the phase angle $\pi/2$.

4.1 Reduction of the amplitude equations

In this Section we perform a standard procedure for the simplification of the amplitude equations (58)–(60). Substituting f from (60), into the other amplitude equations we get

$$\frac{1}{k} \frac{\partial p}{\partial r} = B_1 \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \psi}{\partial r} \right) - (B_2 k^2 + s\rho) \frac{\psi}{r} + s\gamma_1 \mu_1 \frac{\partial \theta}{\partial r}, \quad (62)$$

$$kp = \frac{1}{r} \frac{\partial}{\partial r} \left\{ r \left[B_3 \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \psi}{\partial r} \right) - (B_4 k^2 + s\rho) \frac{\psi}{r} \right] \right\} - s\gamma_1 \mu_2 \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \theta}{\partial r} \right), \quad (63)$$

$$0 = \mu_2 \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \psi}{\partial r} \right) + \mu_1 k^2 \frac{\psi}{r} + \frac{K}{\gamma_1} \frac{\partial}{\partial r} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \theta}{\partial r} \right) - \left(k^2 + \frac{s\gamma_1}{K} \right) \theta \right], \quad (64)$$

where

$$B_1 = \beta_4 - \gamma_1 \mu_1 \mu_2, \quad B_2 = \beta_2 + \gamma_1 \mu_1^2, \quad B_3 = \beta_2 + \gamma_1 \mu_2^2, \quad B_4 = \beta_3 - \gamma_1 \mu_1 \mu_2. \quad (65)$$

and $B_2 > 0, B_3 > 0$ by virtue of (13). Let a new stream function χ be defined as $\psi = r \partial\chi/\partial r$. The orientational ϑ and kinematic ν_i viscosities, as well as other auxiliary functions, are defined by the following relations

$$\vartheta = \frac{K}{\gamma_1}, \quad \nu_i = \frac{B_i}{\rho}, \quad u_i^2 = k^2 + \frac{s}{\nu_i}, \quad w^2 = k^2 + \frac{s}{\vartheta}, \quad \frac{\vartheta}{\nu_i} \ll 1 \rightarrow u_i^2 \leq w^2, \quad (66)$$

where the first inequality in (66) applies to known nematic LC fluids (see Tables 1, 2 in Appendix). Using the new notations we find the first integrals of the amplitude equations as,

$$\frac{p}{k} = (B_1\Delta_{2c} - B_2u_2^2)\chi + s\gamma_1\mu_1\theta, \quad (67)$$

$$kp = (B_3\Delta_{2c} - B_4u_4^2)\Delta_{2c}\chi - s\gamma_1\mu_2\Delta_{2c}\theta, \quad (68)$$

$$0 = (\mu_2\Delta_{2c} + \mu_1k^2)\chi + \vartheta(\Delta_{2c} - w^2)\theta. \quad (69)$$

Next, we eliminate the pressure amplitude p from (67) and (68). This gives,

$$[B_3\Delta_{2c}^2 - (B_1k^2 + B_4u_4^2)\Delta_{2c} + B_2u_2^2k^2]\chi - s\gamma_1(\mu_2\Delta_{2c} + \mu_1k^2)\theta = 0, \quad (70)$$

$$(\mu_2\Delta_{2c} + \mu_1k^2)\chi + \vartheta(\Delta_{2c} - w^2)\theta = 0. \quad (71)$$

Diagonalizing a matrix of operators in (70) and (71) we obtain the following homogeneous equations for the functions $\chi(r)$ and $\theta(r)$,

$$[D_3\Delta_{2c}^3 - D_2\Delta_{2c}^2 + D_1\Delta_{2c} - D_0] \begin{pmatrix} \chi \\ \theta \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}, \quad (72)$$

where

$$\begin{aligned} D_0 &= k^2(\vartheta B_2u_2^2w^2 - s\gamma_1\mu_1^2k^2), \quad D_1 = \vartheta(B_1k^2w^2 + B_2k^2u_2^2 + B_4w^2u_4^2) + 2s\gamma_1\mu_1\mu_2k^2, \\ D_2 &= \vartheta(B_1k^2 + B_3w^2 + B_4u_4^2) - s\gamma_1\mu_2^2, \quad D_3 = \vartheta B_3. \end{aligned} \quad (73)$$

It is easy to verify that all coefficients D_j are positive, if the conditions that all $B_i > 0$ and $\mu_2 \ll 1, \vartheta/\nu_i \ll 1$ are satisfied. The latter are in a good agreement with numerous observations in nematic LCs [13].

Further factorization (recalling that $D_3 > 0$) of the polynomial differential operator gives

$$D_3\Delta_{2c}^3 - D_2\Delta_{2c}^2 + D_1\Delta_{2c} - D_0 = D_3(\Delta_{2c} - m_1^2)(\Delta_{2c} - m_2^2)(\Delta_{2c} - m_3^2). \quad (74)$$

Equation (74) facilitates the following finite solutions of equations (72)

$$\chi(r) = \sum_{j=1}^3 \frac{C_j}{m_j} I_0(m_j r), \quad \theta(r) = \sum_{j=1}^3 \frac{G_j}{m_j} I_0(m_j r), \quad (75)$$

where the second fundamental solutions that diverge at $r = 0$ were excluded, C_j and G_j are indeterminate coefficients and m_j^2 are three generic ⁴ roots of the following cubic equation

$$D_3 m^6 - D_2 m^4 + D_1 m^2 - D_0 = 0 \rightarrow \sum_{j=1}^3 m_j^2 = \frac{D_2}{D_3}, \quad \sum_{j \neq k}^3 m_j^2 m_k^2 = \frac{D_1}{D_3}, \quad \prod_{j=1}^3 m_j^2 = \frac{D_0}{D_3}. \quad (76)$$

The coefficients G_j can be expressed through C_j , once (75) is inserted into (71),

$$G_j = \frac{1}{\vartheta} g_j C_j, \quad g_j = \frac{\mu_1 k^2 + \mu_2 m_j^2}{w^2 - m_j^2}, \quad j = 1, 2, 3. \quad (77)$$

The amplitude of the pressure $p(r)$, the stream function $\psi(r)$ and the displacement of a point on the surface $\varsigma(r_0)$ are easily found from (60), (67), (71) and (77),

$$\begin{aligned} p(r) &= k \sum_{j=1}^3 \frac{l_j}{m_j} C_j I_0(m_j r), \quad l_j = B_1 m_j^2 - B_2 u_2^2 + \frac{s}{\vartheta} \gamma_1 \mu_1 g_j, \\ \psi(r) &= r \sum_{j=1}^3 C_j I_1(m_j r), \quad \varsigma(r_0) = \frac{1}{\vartheta} \frac{1}{k} \sum_{j=1}^3 g_j C_j I_1(m_j r_0), \quad j = 1, 2, 3. \end{aligned} \quad (78)$$

Before proceeding on to the end of this Section, we discuss the distribution of the roots m_j^2 of the cubic equation (76) in the complex plane.

First, m_1^2 is always positive since $D_j > 0$, as mentioned above, and following the Descartes' rule of signs interchange in the sequence of coefficients for real algebraic equations. The other two roots $m_{2,3}^2$ are either positive or complex-conjugate with positive real parts. The last case leads in (75) to Bessel functions of complex arguments. This fact can indicate that the separation of the two groups of functions P_1, ζ and Ψ, Θ, F_r by the $\pi/2$ phase angle, is more elaborate than assumed in (57). Another consequence of the existence of complex-conjugated roots m_j^2 , which is more important from the physical standpoint, is appearance of the imaginary contributions in the dispersion equation. This can lead to the complex value of the growth rate $s = \bar{s} + i\omega$, as its solution, and to the non-steady (oscillatory) evolution of the jet, e.g. $\zeta(z, t) \propto \varsigma(r_0) e^{\bar{s}t} \times e^{i(\omega t + kz)}$, where ω denotes frequency of oscillations.

⁴The freedom to choose the physical parameters of LC seems to admit a degeneration of cubic equation (76), when some of the roots m_j^2 can coincide in different ways. By virtue, such coincidence is not important, since it could occur only at specific wave vectors k^* , which the coefficients D_2, D_1, D_0 are dependent upon. By the other hand, this kind of degeneration might be interesting if k^* is accidentally close to the cut-off wave vector k_d , when the breakage of the LC jet develops.

4.2 Dispersion equation

In what follows we derive the dispersion equation $s = s(kr_0)$, which determines the evolution of Rayleigh instability in LC jets. The revised version of the boundary conditions (61) at $r = r_0$, which utilizes the new stream function $\chi(r)$, reads

$$s \frac{\partial \theta}{\partial r} = k^2 \frac{\partial \chi}{\partial r}, \quad s \gamma_1 \mu_2 \frac{\partial \theta}{\partial r} = B_3 \frac{\partial}{\partial r} \Delta_{2c} \chi + B_5 k^2 \frac{\partial \chi}{\partial r}, \quad \frac{s}{k} p = 2s \beta_1 \frac{\partial^2 \chi}{\partial r^2} + \Gamma \frac{\partial \chi}{\partial r}, \quad (79)$$

where $B_5 = \beta_2 + \gamma_1 \mu_1 \mu_2$. Substituting (75) and (78) into (79), and elimination of the coefficients C_1, C_2, C_3 from the linear equations, leads to a (3×3) -determinant equation

$$\det S_{ij} = 0, \quad (80)$$

where

$$\begin{aligned} S_{1j} &= k^2 - \frac{s}{\vartheta} g_j, & S_{2j} &= B_3 m_j^2 + B_5 k^2 - \frac{s}{\vartheta} \gamma_1 \mu_2 g_j, \\ S_{3j} &= \Gamma - s \left[\frac{l_j}{m_j} \frac{I_0(m_j r_0)}{I_1(m_j r_0)} - 2\beta_1 m_j \frac{I_1'(m_j r_0)}{I_1(m_j r_0)} \right], \end{aligned} \quad (81)$$

and $I_1'(y) = dI_1(y)/dy$. Equation (80) is an implicit form of the exact dispersion relation, which is highly complex and cannot be solved analytically in the general case. Nevertheless, here we can verify the fact, that the cut-off wavelength Λ_d does coincide with Λ_s obtained due to Plateau. Indeed, the cut-off regime corresponds to (79) when $s = 0$ and is satisfied for $\Gamma = 0$, i.e. $\Lambda_d = \Lambda_s$. The implications of equation (80) can be extended further: for the study of different modes of LC flow, including oscillations, and in order to describe asymptotic behaviour of LC jets. This is outside the scope of this paper and will be considered elsewhere. In the next Section, we present a case which facilitates decoupling of hydrodynamic and orientational modes, and consequently the solution of the Rayleigh instability problem in closed form.

5 Decoupling of hydrodynamic and orientational modes

In this Section we discuss a case that renders the dispersion equation (80) solvable. Here we encounter another problem: the elasticity of LC and anisotropy of its viscous properties have the same origin and therefore cannot be managed separately. Nevertheless, we consider the case where the dispersion equation (80) can be simplified. The large number of physical

parameters involved (three viscous moduli, two kinetic coefficients λ and γ_1 , orientational ϑ and kinematic viscosities ν_i , and dimensionless parameter \varkappa) call for such a treatment.

This applies to LC with rod-like molecules ($\lambda \simeq 1$) and low orientational viscosity ϑ

$$\mu_1 \simeq 1, \quad \mu_2 \simeq 0, \quad \vartheta \ll \nu_i, \quad k^2 \ll \frac{s}{\vartheta}, \quad (82)$$

where the first three relations in (82) apply to known nematic LC fluids (see Tables 1, 2 in Appendix). The last inequality in (82) applies to the low-viscosity limit which was considered for the kinematic viscosity in ordinary liquids by Rayleigh [1].

In this case the characteristic equation (76) reduces as follows

$$m^6 - \frac{s}{\vartheta} m^4 + \frac{s}{\vartheta} \left(\mathcal{B} k^2 + \frac{s}{\nu_2} \right) m^2 - \frac{s}{\vartheta} k^2 \left(k^2 + \frac{s}{\nu_2} \right) = 0, \quad \bar{\nu}_i = \frac{\beta_i}{\rho}, \quad \mathcal{B} = \frac{\beta_3 + \beta_4}{\beta_2}. \quad (83)$$

The three roots m_j^2 of equation (76) read

$$2m_{1,2}^2 = \mathcal{B} k^2 + \frac{s}{\nu_2} \pm \sqrt{(\mathcal{B}^2 - 4) k^4 + 2(\mathcal{B} - 2) k^2 \frac{s}{\nu_2} + \left(\frac{s}{\nu_2} \right)^2}, \quad m_3^2 = \frac{s}{\vartheta}. \quad (84)$$

A simple analysis of (84) shows that the dimensionless parameter \mathcal{B} has a critical value 2 that separates two different evolution scenaria of the LC jet. If $\mathcal{B} > 2$ then the both roots m_1^2 and m_2^2 are positive and the capillary instability always appears via trivial bifurcation (steady-state instability). This scenario applies to *MBBA* and *PAA* liquid crystals where $\mathcal{B}_{MBBA} = 5.92$, $\mathcal{B}_{PAA} = 7.11$ (see Tables 1 and 2 in Appendix). In the opposite case, $\mathcal{B} < 2$, one can find the regime where the above roots are complex-conjugates. This gives rise to the oscillatory evolution of the jet which appears via Hopf bifurcation (see Section 4.1).

A significant simplification can be obtained if we assume degeneration of the three viscosities at critical value $\mathcal{B}_* = 2$. Indeed, when the viscous moduli β_j satisfy the relation

$$\mathcal{B}_*(\beta_j) = 2 \quad \longrightarrow \quad 2\beta_1 + \beta_3 = 3\beta_2, \quad (85)$$

the three roots m_j^2 of equation (76) are

$$m_{1*}^2 = k^2, \quad m_{2*}^2 = k^2 + \frac{s}{\nu_2}, \quad m_3^2 = \frac{s}{\vartheta}. \quad (86)$$

Note that (85) cancels the last term in (16). The expressions (86) indicate that the problem was decomposed in two parts, or, in other words, the cross-terms in equations (70), (71)

are dropped. Thus, the first part of the problem is associated with Rayleigh instability as described by

$$(\Delta_{2c} - m_{1*}^2) (\Delta_{2c} - m_{2*}^2) \chi = 0 , \quad (87)$$

with boundary conditions (BC) that account for the elasticity

$$\frac{\partial}{\partial r} \Delta_{2c} \chi + k^2 \frac{\partial \chi}{\partial r} = 0 , \quad \frac{s}{k} p = 2s\beta_1 \frac{\partial^2 \chi}{\partial r^2} + \Gamma \frac{\partial \chi}{\partial r} \quad \text{at } r = r_0 . \quad (88)$$

The second part is associated with an orientational instability of the director field $\mathbf{n}(\mathbf{r}, t)$,

$$(\Delta_{2c} - m_3^2) \theta = 0 , \quad \text{with BC } s \frac{\partial \theta}{\partial r} = k^2 \frac{\partial \chi}{\partial r} \quad \text{at } r = r_0 . \quad (89)$$

The solutions of equations (87) and (89) are

$$\chi(r) = \frac{c_1}{m_{1*}} I_0(m_{1*}r) + \frac{c_2}{m_{2*}} I_0(m_{2*}r) , \quad \theta(r) = \frac{c_3}{m_3} I_0(m_3r) . \quad (90)$$

Hence, using these solutions, the hydrodynamic pressure $p(r)$, stream function $\psi(r)$ and surface displacement $\varsigma(r_0)$ are obtained as

$$p(r) = -c_1 s \rho I_0(m_{1*}r) , \quad \psi(r) = r [c_1 I_1(m_{1*}r) + c_2 I_1(m_{2*}r)] , \quad \varsigma(r_0) = \frac{c_3}{k} I_1(m_3 r_0) ,$$

where here the only indeterminate are c_1 and c_2 , while c_3 can be expressed as their linear combination,

$$c_3 \frac{s}{k^2} = c_1 \frac{I_1(m_{1*}r_0)}{I_1(m_3 r_0)} + c_2 \frac{I_1(m_{2*}r_0)}{I_1(m_3 r_0)} , \quad (91)$$

provided that $s = s(kr_0)$ satisfies the dispersion relation which comes from (88), (90)

$$s^2 + \frac{2\overline{\nu}_1 k^2}{I_0(kr_0)} \left[I_1'(kr_0) - \frac{2km_{2*}}{k^2 + m_{2*}^2} \frac{I_1(kr_0)}{I_1(m_{2*}r_0)} I_1'(m_{2*}r_0) \right] s = \frac{\sigma_0 k}{\rho r_0^2} [1 - k^2 r_0^2 (1 + 2\kappa)] \frac{I_1(kr_0)}{I_0(kr_0)} \frac{m_{2*}^2 - k^2}{m_{2*}^2 + k^2} . \quad (92)$$

If $\kappa = 0$ and $\overline{\nu}_1 = \overline{\nu}_2$, then equation (92) is known as *Weber equation* for a viscous isotropic liquid [6]. For low viscosity, $\beta_1 \sim \beta_2 \ll \sqrt{\rho \sigma_0 r_0}$, a Rayleigh type expression is obtained (see Figure 2)

$$s_-^2(kr_0) = \frac{\sigma_0 k}{\rho r_0^2} [1 - k^2 r_0^2 (1 + 2\kappa)] \frac{I_1(kr_0)}{I_0(kr_0)} , \quad (93)$$

where subscript "−" denotes low viscosity.

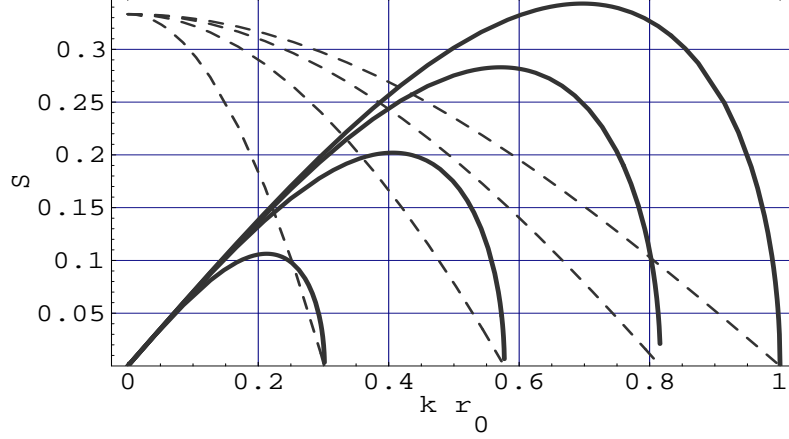


Figure 2: A plot of rescaled growth rate S vs. kr_0 for low viscosity $\sqrt{\rho r_0^3/\sigma_0} s_-(kr_0)$ (plain line) and high viscosity $2\beta_2 r_0/\sigma_0 s_+(kr_0)$ (dashed line) for different values of \varkappa in descending order from above: $\varkappa = 0, 0.25, 1, 5$. If $\vartheta/\nu = 4\varkappa$, then the scaling for both viscous regimes is the same.

The maximum s_-^{max} in equation (93) which corresponds to the wave number k_-^{max} , gives rise to evolution of the largest capillary instability. Numerical calculation shows that s_-^{max} and k_-^{max} are both proportional to $(1 + 2\varkappa)^{-1/2}$

$$s_-^{max} \cong \frac{1}{3\sqrt{1+2\varkappa}} \sqrt{\frac{\sigma_0}{\rho r_0^3}}, \quad k_-^{max} \cong \frac{a}{r_0 \sqrt{1+2\varkappa}}, \quad a = 0.697. \quad (94)$$

When high viscosity prevails $\beta_1 \sim \beta_2 \gg \sqrt{\rho \sigma_0 r_0}$, the dispersion equation reads (see Figure 2)

$$s_+(kr_0) = \frac{\sigma_0}{2\beta_2 r_0^2 k} \frac{[1 - k^2 r_0^2 (1 + 2\varkappa)] I_1^2(kr_0)}{I_0(kr_0) I_1(kr_0) + kr_0 [I_1'(kr_0)]^2}, \quad s_+^{max} \cong \frac{\sigma_0}{6\beta_2 r_0}, \quad k_+^{max} = 0. \quad (95)$$

where subscript “+” denotes high viscosity. Similar to ordinary liquids [4], in this limit there is no finite mode of maximum instability for any \varkappa . In this case we have

$$\varsigma(r_0) = \frac{k_+^{max}}{s_+^{max}} [c_1 I_1(k_+^{max} r_0) + c_2 I_1(m_{2*} r_0)] = 0. \quad (96)$$

Nevertheless, there exists a continuous range $[0, (1 + 2\varkappa)^{-1/2} r_0^{-1}]$ of wave numbers k , with finite disturbance growth rate $s_+(kr_0)$, which affect the cylindrical jet.

5.1 Hydrodynamic influence on LC’s orientational instability

We conclude this Section with a brief discussion regarding the hydrodynamic influence on the orientational instability of the director field $\mathbf{n}(\mathbf{r}, t)$. As the effect of hydrodynamics changes

the wave number k_s of Plateau instability to k_{max} , the flow drives the orientational instability (43) of the director field $\mathbf{n}(\mathbf{r}, t)$. Indeed, according to (90)

$$n_r^1(r, z) = c_3 I_1(m_3^{max} r) , \quad m_3^{max} = \sqrt{\frac{s^{max}}{\vartheta}} . \quad (97)$$

It is convenient to consider the following two marginal viscous regimes.

1. The low-viscosity limit:

$$(m_{3-}^{max} r_0)^2 \cong \frac{1}{3\sqrt{1+2\kappa}} \frac{1}{\sqrt{\kappa\varepsilon}} , \quad \varepsilon = \frac{\rho K}{\gamma_1^2} , \quad (98)$$

where $\varepsilon \sim 10^{-6} \div 10^{-4}$ is a small dimensionless parameter.

2. The high-viscosity limit:

$$(w_{3+}^{max} r_0)^2 \cong \frac{1}{6\kappa} \frac{\gamma_1}{\beta_2} . \quad (99)$$

In both limits the distribution of director field $\mathbf{n}(\mathbf{r}, t)$ in the jet is always nontrivial and definitely far from static distribution (43).

6 Conclusion

1. The capillary instability of liquid crystalline (LC) jet is considered in the framework of linear hydrodynamics of uniaxial nematic LC. Its static version, called Plateau instability and being correspondent to the variational problem of minimal free energy, predicts an essential dependence of the disturbance *cut-off* wavelength upon the dimensionless parameter $\kappa = K/\sigma_0 r_0$.

2. The hydrodynamic problem of capillary instability in LC jets is solved exactly followed by derivation of the dispersion relation. This relation, which is represented as a determinant equation, expresses implicitly the dispersion $s = s(k)$ of the growth rate s , as a function of the wave number k of axisymmetric disturbances of the jet.

3. The case, where the dispersion equation becomes solvable, is considered in detail. It corresponds to the regime, wherein the hydrodynamic and orientational modes become decoupled. Hydrodynamics changes the wave number k_s of Plateau instability into k_{max} that produces evolution of the largest capillary instability. Similarly, hydrodynamic flow influences the static orientational instability of the director field $\mathbf{n}(\mathbf{r}, t)$.

4. The present theory can be easily extended to non-uniaxial nematic LC, which possesses finite point symmetry groups $G \subset O(3)$ as distinguished from uniaxial group $D_{\infty h}$. The corresponding expressions for the free energy density $E_d(G)$ and the dissipative function $D(G)$ were derived in [21].

5. In this work the effect of external fields was not considered. However, the theory developed here facilitates the treatment of Rayleigh instability in nematic LC in the presence of static electromagnetic fields.

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A Appendix

Table 1. The basic physical parameters α_i , ρ , K , σ_0 and their derivatives η_i , β_i , γ_i , B_i , μ_i , λ and ν_i for nematic liquid crystal *4-metoxybenziliden-4-butilanilin (MBBA)* at 25°C taken from [19], [20].

$\alpha_1, mPa \cdot s$	$\alpha_2, mPa \cdot s$	$\alpha_3, mPa \cdot s$	$\alpha_4, mPa \cdot s$	$\alpha_5, mPa \cdot s$	$\alpha_6, mPa \cdot s$
7	-78	-1	84	46	-33
$\eta_1, mPa \cdot s$	$\eta_3, mPa \cdot s$	$\eta_5, mPa \cdot s$	λ	μ_1	μ_2
42	50	104	1.026	1.013	0.013
$\beta_1, mPa \cdot s$	$\beta_2, mPa \cdot s$	$\beta_3, mPa \cdot s$	$\beta_4, mPa \cdot s$	$\gamma_1, mPa \cdot s$	$\gamma_2, mPa \cdot s$
42	25	79	59	77	-79
$B_1, mPa \cdot s$	$B_2, mPa \cdot s$	$B_3, mPa \cdot s$	$B_4, mPa \cdot s$	\mathcal{B}	$\vartheta, m^2/s$
58	104	25	78	5.92	1.2×10^{-10}
$\rho, kg/m^3$	K, N	$\sigma_0, N/m$	$K/\sigma_0, m$	$\nu_i, m^2/s$	ϑ/ν_i
1.2×10^3	9×10^{-12}	38×10^{-3}	2.4×10^{-10}	$10^{-5} \div 10^{-4}$	$10^{-6} \div 10^{-5}$

Table 2. The basic physical parameters α_i , ρ , K , σ_0 and their derivatives η_i , β_i , γ_i , B_i , μ_i , λ and ν_i for nematic liquid crystal *para-azoxyanisole (PAA)* at 122°C taken from [19], [20].

$\alpha_1, mPa \cdot s$	$\alpha_2, mPa \cdot s$	$\alpha_3, mPa \cdot s$	$\alpha_4, mPa \cdot s$	$\alpha_5, mPa \cdot s$	$\alpha_6, mPa \cdot s$
4	-6.9	-0.2	6.8	5	-2.1
$\eta_1, mPa \cdot s$	$\eta_3, mPa \cdot s$	$\eta_5, mPa \cdot s$	λ	μ_1	μ_2
3.4	4.5	13.7	1.06	1.03	0.03
$\beta_1, mPa \cdot s$	$\beta_2, mPa \cdot s$	$\beta_3, mPa \cdot s$	$\beta_4, mPa \cdot s$	$\gamma_1, mPa \cdot s$	$\gamma_2, mPa \cdot s$
3.4	2.25	11.45	4.55	6.7	-7.1
$B_1, mPa \cdot s$	$B_2, mPa \cdot s$	$B_3, mPa \cdot s$	$B_4, mPa \cdot s$	\mathcal{B}	$\vartheta, m^2/s$
4.34	9.36	2.26	11.24	7.11	1.8×10^{-9}
$\rho, kg/m^3$	K, N	$\sigma_0, N/m$	$K/\sigma_0, m$	$\nu_i, m^2/s$	ϑ/ν_i
1.4×10^3	11.9×10^{-12}	40×10^{-3}	3×10^{-10}	$10^{-6} \div 10^{-5}$	$10^{-4} \div 10^{-3}$

